

## A Review on Carbonation of Concrete and Its Predictive Modelling

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### Abstract

The phenomenon of concrete carbonation is a well-researched field. With ever rising concern of pollution in environment, need for understanding this phenomenon has increased many folds. Although, there is plenty of experimental data is an available, useful and quantifiable generalisation of influences of different factors affecting parameters is a difficult task as there is innumerable variability in experimental conditions and ingredients of the concretes tested and the specimens taken for conduction of tests. Several studies have employed numerical and statistical modelling techniques to predict the depth of carbonation as a function of the age of concrete, compressive strength of concrete and other parameters affecting carbonation of concrete. This paper discusses several issues of uncertainty in prediction and evaluation of carbonation propagation.

**Keywords:** Carbonation, microstructure of concrete; Service life, durability.

### 1. INTRODUCTION

The physical, mechanical and chemical properties of both components of Reinforced Concrete Structures (i.e. steel and concrete) improves by their synergy. The steel is protected by concrete layer from various physical and chemical agents, which solves the major problem that affects metallic structures, namely the corrosion of steel when exposed to the environment. (Parrott, 1987; Kulakowski *et al.* 2009)

Corrosion of reinforcement is repressed to some extent due to high pH of concrete, which is around pH 12 to 13. A protective oxide layer is formed on the surface of steel which impedes metals from dissolving. However, this protective layer is destroyed when alkalinity of encompassing concrete gets reduced. (Parrott 1987; Broomfield 2006; Khan and Lynsdale 2002). Ingress of aggressive materials like carbon dioxide and chloride ions takes place through the random porous structure of concrete. Due to the carbonation reaction, taking place between carbon dioxide and hydration products of cement, carbonates are formed which causes the reduction in alkalinity of concrete. The reduction in alkalinity also reduces concrete's ability to protect the steel reinforcement from corrosion (Verbeck, 1975; Parrott, 1992; Currie, 1986; Ho and Lewis 1987; Wierig, 1984). Hobbs (Hobbs 1988) suggested that the threshold value of pH for steel de-passivation is 9.5 below which the steel corrosion takes place

Because carbonates formed occupies a greater volume than hydroxides which it replaces, the porosity of carbonated concrete is reduced. Also, water released by hydroxides on carbonation may aid the hydration of un-hydrated cement. These changes result in increased surface hardness, increased strength at the surface (Baba and Senbu 1987), reduced surface permeability, reduced moisture movement (Papadakis *et al.* 1991) and increased resistance to those forms of deteriorating attacks which are controlled by porosity (Burkan Isgor and Razaqpur 2004). It is well known that carbonation also causes an increase in all mechanical properties of concrete like strength, modulus of elasticity and shrinkage of the concrete (Chang *et al.* 2003).

#### 1.1. Mechanism of Carbonation

The process of carbonation of concrete is a combination of various physical and chemical processes. The carbon dioxide is present in ambient atmosphere or internal air of buildings. The instrumental mechanism of carbonation is the reaction of diffused CO<sub>2</sub> with calcium hydroxide. The final products of this reaction are CaCO<sub>3</sub> and water.

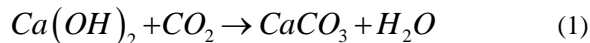
- (1) The physicochemical processes involved in carbonation are: (Thiery *et al.* 2007)
- (2) The diffusion of CO<sub>2</sub> in the gaseous phase of the concrete pores.

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- (3) The dissolution of  $\text{CO}_2$  in the pore water as carbonic acid.
- (4) Dissociation of  $\text{CO}_2$  as  $\text{H}_2\text{CO}_3$  and  $\text{CO}_3^{2-}$  ions.
- (5) The dissolution of solid  $\text{Ca}(\text{OH})_2$ .
- (6) Releasing of calcium  $\text{Ca}^{2+}$  and hydroxyl  $\text{OH}^-$  ions.
- (7) And the precipitation of  $\text{Ca}^{2+}$  with  $\text{CO}_3^{2-}$ , forming  $\text{CaCO}_3$

These intermediate reactions are represented in the following overall chemical reaction given in equation 1:



Apart from  $\text{Ca}(\text{OH})_2$ , several other hydration compounds also react with  $\text{CO}_2$ . The reaction between  $\text{CO}_2$  and the calcium silicate hydrates (C-S-H) produces  $\text{CaCO}_3$  and a silica gel. (Šauman, 1971; Slegers and Rouxhet 1976; Kobayashi *et al.* 1994) In the case of a rapid-hardening cement with a high  $\text{C}_3\text{A}$  content, or a cement mixed with a large amount of gypsum, Ettringite (tri-sulfate hydrates AFt) and mono-sulfate hydrates (AFm) are also formed in abundance (Thiery *et al.* 2007).

## 1.2 Carbonation: A Nonlinear Phenomena

Many studies have been carried out on carbonation of Concrete. However, the generalisation of observed trends seems to be an almost impossible task as there are innumerable variations in experimental conditions and compositions of the concretes tested.

The above-mentioned processes (section 1.1) are governed by their own factors, which are interdependent. For example, diffusion of  $\text{CO}_2$  is dependent upon transport properties of concrete structure. These transport properties are governed by moisture content, type of cement and temperature of the media. As seen in chemical reaction, water is formed at the end the reaction. This will affect the diffusion of  $\text{CO}_2$  and water (Lagerblad, 2005). Because of these coupling or synergic effects of influencing parameters involved at almost every step of the process, any analytical model for prediction of carbonation is inherently complex and should be based on a non-linear scheme.

For reinforced concrete structures, the onset of reinforcement corrosion is considered as one of the critical stages for determining the service life of a concrete structure (Khunthongkeaw *et al.* 2006). Carbonation is a process from the surface, i.e. the amount of carbonated material is related to exposure time and surface. Surfaces in direct contact with carbon dioxide and water will carbonate rapidly, but a shell of already carbonated concrete will slow down the carbonation of the interior. The rate of carbonation is

not constant. The carbonation will eventually slow down as the carbon dioxide must pass through a thickening layer of concrete's altered products (Lagerblad, 2005).

## 2. OVERVIEW OF SOME CARBONATION MODELS PROPOSED

Based on many experimental studies in the past along with analytical hypothesis proposed by many authors, several mathematical models have been developed to predict the concrete service life of the structure.

Several early experimental studies on concrete carbonation, for outdoor conditions (Shigeyoshi Nagataki and Eun Kyum Kim 1986) and for controlled indoor conditions (Tuutti 1982; Shigeyoshi Nagataki and Eun Kyum Kim 1986; S. Nagataki and H. Ohga 1988; Qiu-Dong 1987) hypothesises that depth of concrete carbonation or the location of "carbonation front" is directly proportional to square root of age of concrete.

i.e.  $\sqrt{t}$  (Papadakis Vayenas, *et al.* 1991) The expression used is given in equation 2.

$$X_c = A\sqrt{t} \quad (2)$$

Where,  $X_c$  = carbonation depth after time  $t$ ,  $t$  = carbonation exposure duration, and  $A$  = empirical constant. In this model, it is to be noted that variable nature of the diffusivity constant of  $\text{CO}_2$  was not considered (Houst and Wittmann 2002). In reality, the value of  $A$  should depend upon the degree of saturation of concrete, the pore structure of concrete, and the carbon dioxide content of the environment.

A more accurate model, based on the microstructure of concrete, which is quite similar to equation 2 was developed by (Papadakis Vayenas *et al.* 1991). This mathematical model yielded a complex nonlinear system of differential equations in space and time and had to be solved numerically for the unknown concentrations of the materials involved (Papadakis Fardis *et al.* 1991). This model predicted the Equation 3, for giving depth of carbonation.

$$X_c = \sqrt{\frac{2D_{e,\text{CO}_2}^C [\text{CO}_2]^0}{[\text{Ca}(\text{OH})_2]^0 + 3[\text{CSH}]^0 + 3[\text{C}_3\text{S}]^0 + 2[\text{C}_2\text{S}]^0}} t \quad (3)$$

Where,  $D_{e,\text{CO}_2}^C$  = effective diffusivity of  $\text{CO}_2$ ,  $[\text{Ca}(\text{OH})_2]^0$ ,  $[\text{Ca}(\text{OH})_2(\text{s})]^0$ ,  $[\text{CSH}]^0$ ,  $[\text{C}_3\text{S}]^0$ , and  $[\text{C}_2\text{S}]^0$  are the "initial" concentrations (at  $t=0$ ) of  $\text{Ca}(\text{OH})_2(\text{aq})$ ,  $\text{Ca}(\text{OH})_2(\text{s})$ , CSH,  $\text{C}_3\text{S}$ , and  $\text{C}_2\text{S}$  respectively at the end of moist curing. In this model, carbonation effects of anhydrous cement clinker phases, concrete porosity and degree of saturation were also taken into account. The proposed model could predict the test results of previous researchers, for exposure to natural or higher concentrations of ambient  $\text{CO}_2$ .

There are other several statistical and linear regression-based models available for the relationship among influencing factors, carbonation depth and strength (Taffese *et al.* 2015; Atiş 2004; Leemann *et al.* 2015; Hills *et al.* 2015; J.-K. Kim and Lee, 1999).

Apart from empirical models, several finite element based models (Martin-Peréz *et al.* 1998; Burkan Isgor and Razaqpur 2004; H.-W. Song *et al.* 2006; Hussain 2011) also has been used to predict the temporal variation of carbonation depth. A nonlinear finite element approach for tracing the spatial and temporal advancement of the carbonation front in concrete structures with and without cracks was developed by (Burkan Isgor and Razaqpur 2004). A two-dimensional Windows-based finite element computer program, called CONDUR, was developed. The diffusion of carbon dioxide into concrete was modelled as an analogy with two-dimensional transient heat transfer problem. The dependency of carbon dioxide diffusion rate on the moisture and the temperature was considered. Similarly, moisture transport was also simulated. The progress of carbonation reaction at each time step was determined using chemical kinetics data available. A coupling mechanism between all these processes was achieved using source/sink terms in each process. The relative humidity or the moisture present and concentration of  $CO_2$  in each element was updated at each time step. To determine the onset of rebar corrosion, the time-dependent reduction in pH due to carbonation was determined by using data related to progress of carbonation reaction (equation 4). The results obtained from the program were compared with available experimental data.

More recently, a Finite elemental framework developed by Behrouz Shafei (Shafei *et al.* 2012), evaluates the effects of the most critical parameters that may accelerate or slow down the carbonation process. The analyses of parameters are also based on the analogy of several processes involved in concrete carbonation with transient thermal analysis with appropriate modifications. The novelty of the proposed framework was to consider the nonlinear time-dependent characteristics of the involved parameters along with their mutual interactions in ANSYS software.

Artificial neural networks and other machine learning techniques have also been used (Liu *et al.* 2008; Kwon and Song 2010; Taffese *et al.* 2015). As it is very difficult to establish a universal carbonation model which can represent all factors because of the numerous influencing factors of carbonation and large data discretisation in the laboratory testing or actual site. The artificial neural networks can be very efficiently utilised for this purpose (Liu *et al.* 2008).

A Radial Basis Function (RBF) neural network based model has been established by (Liu *et al.* 2008).

Water-gel ratio, cement content and time of exposure of concrete were selected as input parameters in the study, while output layer had only one node, i.e. carbonation depth of concrete. 72 groups of carbonation data were used in building RBF and BP neural network model, among which 60 groups of data were used to train, and another 12 groups of data were used for validation of the model. The prediction results show that the forecast results conform to the test results very well. Through the comparison of results, it was concluded that the RBF neural network is more superior than BP network in both identifying effect and evaluation precision.

Apart from actual carbonation depth, ANNs can be very useful in the determination of several other parameters like diffusion coefficient etc. In the study (Kwon and Song 2010), different experimental results were analysed to obtain the comparable data set of the  $CO_2$  diffusion coefficient. Several mix design components (i.e. cement content, water to cement ratio, and volume of aggregate including exposure condition of relative humidity) were selected as neurons followed by training of learning for an eural network using backpropagation algorithm. The diffusion coefficients of  $CO_2$  obtained from the neural network were found to be in good agreement with experimental data considering various conditions. Finally, the numerical technique which is based on behavior in early- aged concrete such as hydration and pore structure was developed considering  $CO_2$  diffusion coefficient from neural network and changing effect on porosity under carbonation.

### 3. DISCUSSION: EXPERIMENTAL STUDIES AND PREDICTION MODELS FOR DIFFERENT INFLUENCING FACTORS

A need to review the already existing findings and facts is evolved, to help in paving the way for further investigations. So, the effects of the several factors affecting carbonation and environmental loading which affect the service life of concrete structures are described in detail. The models proposed and issues of uncertainty in prediction and evaluation of carbonation propagation are also discussed in this section.

#### 3.1 Effect of Curing Condition

The influence of curing on concrete carbonation can be in terms of the type of curing and in terms of initial curing period. Conclusions drawn upon Impact of curing may also be different for natural long-term carbonation with site-curing and accelerated carbonation with laboratory-controlled curing. In a critical review by (Ekolu, 2016), the author points out that the studies on effects of different curing methods on natural carbonation of concrete in real structures are very rare. Different methods of curing have been studied in the laboratory including heat curing (Neville, 1995; Ekolu, 2006; Lo *et al.* 2016), air curing,

moistcuring (Neville, 1995; Lo and Lee, 2002; Aprianti et al. 2016; Balayssac et al. 1995) and use of curing compounds (Neville, 1995; Xue et al. 2015).

In an experimental study, (Lo and Lee, 2002) prepared three different normal weight concrete grades A, B and C with water-cement ratios of 0.38, 0.46 and 0.54, respectively, to study the effects of initial curing method on the depth of carbonation. The details of 3 mixes are given in Table 1.

**Table 1. Mix Proportion Per Cubic Meter of Concrete**

Properties	Mix A	Mix B	Mix C
w/c Ratio	0.38	0.46	0.54
OPC(kg)	450	450	450
20 mm granite aggregate (kg)	665	655	645
10 mm granite aggregate (kg)	530	515	510
Rock fines (kg)	535	525	515
Superplasticizer (ml)	2400	0	0

For each grade, 8 cubes of 100×100 mm<sup>2</sup> dimension and 8 units of 100Ø×200 mm high cylinders were cast and stored in water at 27 ± 3°. After 28 days, the cubes were tested for compressive strength. The cylinders were taken out from the carbonation chamber after 30, 60, and 90 days and were split in a tensile splitting test. The transverse exposed surfaces were cleaned and tested for carbonation depth using phenolphthalein indicator and a Fourier-transform infrared spectroscopy.

It was observed that the carbonation depth increases with w/c ratio, the age of concrete and the initial curing period. This difference in carbonation depths between air cured and water cured samples were very small with moist curing giving slightly lower carbonation depth. The discrepancy in the carbonation depth measured by IR for both air- and water-cured conditions reduced over time. Samples cured in water for 28 days became carbonated to only 53% of the level for air-cured samples but then increased to 78% after the three-month period (Lo and Lee, 2002). It is also to be noted that 28-days strengths of moist cured samples were slightly higher.

In a critical review on effect of curing on long-term natural carbonation of concrete, (Ekolu, 2016) cited the long-term experimental studies by CSIR (Xue et al. 2015) and highlighted the fact that, none of the curing regimes caused a change of strength class of a given mixture or any change in their respective carbonation behaviors under outdoor exposure, refer table 2. In this study, it was observed that, for each strength class, the carbonation depths at later ages differ by about ±2 mm, regardless of the curing regime. Statistical analysis was conducted to assess whether the concretes subjected to the different curing regimes exhibited a significant change in carbonation. 95% confidence limit graphs were plotted. The residuals fell

within the 95% confidence interval, indicating the absence of statistically significant differences between results of the various curing regimes. It was concluded that it is only if a curing method causes a change in strength class of a mixture, that corresponding carbonation behaviour of the mix can be expected to alter significantly.

**Table 2. Results of Carbonation Depth Using Phenolphthalein Test And Infrared Spectroscopic Test**

Mix (w/c)	Curing type	Strength (MPa)	Carbonation depth (mm)		
			Phenolphthalein P infrared spectrum(I)		
Age (month)			1	2	3
A (0.38)	Air	66	1 (1.5)	3.5 (4.5)	5 (6)
	Water	78	0.5 (1)	1.5 (3)	3 (4.5)
B (0.46)	Air	44	1.5 (3)	4 (6)	7 (8)
	Water	63	1 (3)	2.5 (4.5)	5 (6)
C (0.54)	Air	35	3 (4.5)	5.5 (8)	10 (12)
	Water	54	1.5 (3)	4 (6)	8.5 (10)

**Table 3. Properties Of Concrete Used By (Balayssac et al. 1995)**

Concrete	Cement	cement content (C) (kg m <sup>-3</sup> )	Water/Cement	Gravel/Sand	Characteristic Strength (MPa)
B1	CPJ45	300	0.65	1.00	25.1
B2	CPJ45	340	0.61	1.13	32.6
B3	CPJ45	380	0.53	1.13	37.8
B4	CPJ45	420	0.48	1.15	43.5
-	CPA55	250	0.73	0.92	26.4
-	CPA55	280	0.65	0.94	30.0
-	CPA55	300	0.59	0.96	35.0
-	CPA55	350	0.54	1.04	41.8
-	CLK45	340	0.61	1.17	24.9
-	CLK45	400	0.50	1.15	31.9

The initial curing period of concrete plays a significant role in carbonation of concrete. When curing is conducted for the proper time, the susceptibility of concrete to carbonation reduces. The purpose of curing is to maintain a required range of moisture and temperature.

An experimental study was conducted by (Balayssac et al. 1995) using concrete mixes of a range of strength from 25 to 40 MPa. All mixes provided the



same slump (8 cm). Cement content, w/c ratios and 28-day strengths are given in Table 3 and Oxide composition and physical properties of cement are given in Table 4.

**Table 4. Oxide Composition And Physical Properties Of Cement Used In (Balayssac et al. 1995)**

Cement	CPJ45	CPA55	CLK45
SiO <sub>2</sub>	26.2	20.1	30.3
Al <sub>2</sub> O <sub>3</sub>	3.1	5.4	11.5
Fe <sub>2</sub> O <sub>3</sub>	3.2	3.2	-
CaO	58.9	63.2	45.2
MgO	0.6	2.2	4.75
SO <sub>3</sub>	2.6	2.3	2.9
K <sub>2</sub> O	-	1.22	0.76
Na <sub>2</sub> O	-	-	0.6
Ss(m <sup>2</sup> kg <sup>-1</sup> )	350	370	385
Density	3.01	3.15	2.93

After demoulding at one day, the test samples were stored in a controlled environment (20°C and 60% relative humidity) or kept in water up to 3 or 28 days, before exposure to the environment (with a CO<sub>2</sub> content of 0.03%), for up to 18 months. Cylinders with 11 cm diameter and 10 cm height were used in this study whose ends were sawn after demoulding. Also, after curing the lateral surfaces were covered with aluminium sheets. After an appointed period (90, 180, 360 and 540 days) three test samples were split to obtain a break without wet sawing. The carbonated depth was measured using phenolphthalein solution.

Carbonation depth decreased with increase in cement content for any curing time, (also 28-day strength). For the concrete with a cement content of 350 kg m<sup>-3</sup>, increasing the period of curing from 1 days to 28 days caused the reduction of the carbonation depth to half. When the curing period was increased from 1 to 3 days, there was a rapid decrease in carbonation depth followed by slower variations. It is to be noted if cement

content is too low, carbonation depth is still high in spite of good curing. Also, curing effects depend on cement type. Increasing curing period does not have the same effect, for the same cement content, on the concrete of Portland Cement as on the concrete of slag cement. One day curing was found to be insufficient for all the concretes regardless of the cement content. In conclusion, curing period of 3 days is sufficient for concretes with a cement content higher than 380 kg m<sup>-3</sup> for good carbonation resistance. Detailed data on carbonation depths obtained are given in Table 5.

Although, some statistical and machine learning based models are present (Taffese et al. 2015; Hills et al. 2015; Kwon and Song 2010; J.-K. Kim and Lee, 1999), analytical models involving the curing process were found to be very rare. The FIB- model (FIB 2006) accounts for the effect of site curing by providing a correction factor, which is dependent on the length of curing but it does not mention anything on implications of different curing methods on natural carbonation. (Ekolu, 2016).

### 3.2 Ambient Temperature and Thermal Properties

Small variations in temperature have little effect on carbonation. High temperature increases the carbonation rate unless drying overcomes the temperature effect. (Neville, 1995). It also affects the passivity of steel rebars in reinforced concrete. Although, the thermal conductivity of concrete is a well-researched topic because of its importance for fire resistant design and energy conservation, its variation during carbonation of cement materials has not been studied to a good extent. The heat of hydration of concrete or occurrence of temperature gradient may lead to cracks in a concrete structure. (Kim et al. 2003) Propagation of cracks increases the susceptibility of concrete to carbonation.

**Table 5. Carbonation Depths Observed In (Balayssac et al. 1995)**

Concrete	Properties	Curing Period	Age(day)			
			90	180	360	540
B1	Cement content=300 kg m <sup>-3</sup> W/C=0.65 28-day strength=25.1 MPa	1 day	6.5	11	13	15
		3 day	4	6	9.5	13
		28 day	3	5	6	9
B2	Cement content=340 kg m <sup>-3</sup> W/C=0.61 28-day strength=32.6 MPa	1 day	5.5	10	12	13
		3 day	3.5	5	6	8
		28 day	2.5	4	4.5	6
B3	Cement content=380 kg m <sup>-3</sup> w/c=0.53 28-day strength=37.8 MPa	1 day	4.5	9	10	11.5
		3 day	3	5	5.5	7
		28 day	2	3.5	4	5
B4	Cement content=420 kg m <sup>-3</sup> w/c=0.48 28-day strength=43.8 MPa	1 day	4	7.5	8.5	9.5
		3 day	2.5	4	3.5	4
		28 day	1.5	3	3	3.5

### 3.2.1 Effects of Ambient Temperature on Passivity Capacity of Reinforcement

Jiezhen (Hu *et al.* 2015) studied the coupled effect of temperature and carbonation on the corrosion of reinforcement by using open circuit method (OCP), the electrochemical impedance spectroscopy (EIS) and the potentio-dynamic polarisation (PP) in the simulated concrete pore solutions (SPSs). The OCP was used to evaluate corrosion tendency of rebar in SPSs, the passivation film of the rebars was evaluated by using EIS and the corrosion current or dissolution velocity of the passivated surface of the rebars was evaluated using potentio-dynamic polarisation measurements.

Following observations were made:

- The Effect of Temperature and pH on the OCP: The rebars were found to be prone to corrosion in higher temperature and lower pH SPSs. The corrosion of rebar was high at the pH = 9.6 and 318K SPSs. The rebars had the best corrosion-resistance at the pH= 10.6 and 318K temperature of SPSs.
- The Effect of Temperature and pH on the Passivation Film of Rebar: The rebars were found to have a greater capacity of passivity at a lower temperature. Moderate pH value (11.6) SPSs was found to be favourable for passivation layer.
- The Effect of Temperature and pH on the Corrosion Current of Rebar: The passive current of rebars in the higher pH value SPSs was bigger than that of rebars in lower pH value SPSs. This means that the dissolution velocity of the passivated surface is bigger if the pH of the SPSs is very high. Also, the high temperature and high pH environment are favourable for the formation of passivated surface.

In conclusion, the high-temperature environment is favourable for the formation of the passivated surface of rebars in the SPSs. But the dissolution velocity of the passivated surface is higher in the high-temperature SPSs. Also, rebars have a greater capacity of passivity at a lower temperature. The corrosion rate of rebars at higher temperature is smaller in moderate pH value (10.6) SPSs. The rebars suffer from serious corrosion in the pH= 9.6 SPSs at (High) 318K temperature.

For a service life model for Reinforced Concrete, it is very important to monitor the change in the pH of the pore solution. As discussed above, the onset of corrosion as well as the degree of corrosion is highly dependent on pH of pore solution. As per (Papadakis, Fardis, *et al.* 1991) the pH of carbonated concrete can be written as equation 4 :

$$pH = 14 + \log \left( 2 \times 10^3 \right) \left[ Ca(OH)_2 \right]_{aq} \quad (4)$$

As per the best knowledge of the authors, most of the models available for service life prediction of reinforced concrete doesn't consider the effect of temperature on passivity of rebars. The threshold value pH for onset of corrosion is dependent upon the temperature.

### 3.2.2 Effects of Sudden Changes in Temperature

S.K. Roy (Roy *et al.* 1999) explains that concrete structures which are exposed to the environment, faces not only seasonal and long-term changes in temperature but also face sudden changes due to rain or due to change in temperature during day and night cycle. When there is sudden fall in temperature, the outer concrete surface contracts more than the inside and tensile stress occur at the surface and compressive stress inside. Also, while the temperature rises suddenly, the outer surface of concrete expands more than the inside and compressive stresses develop at the surface and tensile stresses develop inside. This may lead to the formation of microcracks in concrete structures increasing the surface are exposed to CO<sub>2</sub>. Leading to an increase in carbonation.

### 3.2.3 Effects on Thermal Conductivity

There is a good agreement to the fact that carbonation causes a reduction in the total porosity of concrete and alters its pore size distribution. The overall effect is that carbonation tends to increase the thermal conductivity appreciably. (Pham and Prince 2013).

The thermal conductivity of concrete is a function of mix properties, types of aggregates used, as well as moisture status. (Morabito, 1989); (Neville, 1995) and (Lanciani *et al.* 1989). Higher w/c ratios give a more porous structure. The pores have low thermal conductivity and heat capacity compared to the other phases present in concrete. More porosity enables a better scope for the influence of moisture content on the heat capacity of concrete. The moisture in any form in concrete, i.e. chemically bound, physically bound or free water, have higher heat capacity.

The changes in the microstructure and macrophysical properties caused by the carbonation of normalised CEM II mortar were examined in detail by (Pham and Prince, 2013). Normalized mortar prepared with French Cement CEM II / BM (V-LL) 32.5 R and standard sand certified in accordance with EN 196-1 were used. The w/c and sand/cement ratios were respectively 0.5 and 3. At the end of the mixing, the mortar was placed in cylindrical moulds (Diameter = 40mm, height = 60mm). The samples were demoulded after 24 hours and then cured for 90 days in a humid chamber (20°C, 100% relative humidity). Samples were subjected to accelerated carbonation at 20°C, 65% relative humidity and 20% CO<sub>2</sub> concentration. A Hot Disk Thermal Constants analyser was used to perform the thermal conductivity test at 20°. A plane Hot Disk sensor was fixed between two pieces of the sample – each one with a plane surface facing the sensor. The hot

Disk sensor had a dynamic role. The disc was used both as a heat source and as a dynamic temperature sensor. Electrical current, high enough to increase the temperature of the sensor was passed. The increase in resistance (temperature) was recorded as a function of time. Fig. 1 presents the variation of thermal conductivity during carbonation. The study confirmed that the thermal conductivity increases with increase in carbonation.

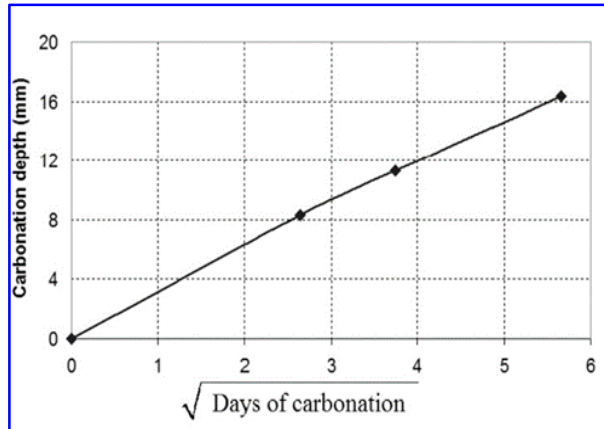


Fig. 1: Evolution of thermal conductivity with carbonation; Source: (Pham and Prince 2013)

### 3.2.4 Modeling of heat flow in concrete

The modelling of heat flow inside concrete can be done using Fourier's law, (Burkan Isgor *et al.* 2004) which is given by equation 5.

$$-\frac{\partial}{\partial x}\left(-k \frac{\partial T}{\partial x}\right) - \frac{\partial}{\partial y}\left(-k \frac{\partial T}{\partial y}\right) + Q = \rho c \frac{\partial T}{\partial t} \quad (5)$$

Where,  $k$  is the thermal conductivity of concrete ( $\text{Wm}^{-1} \text{ } ^\circ\text{C}$ ),  $T$  is its temperature ( $^\circ\text{C}$ ),  $\rho$  is its density ( $\text{kg m}^{-3}$ ),  $c$  is its specific heat ( $\text{Jkg}^{-1} \text{ } ^\circ\text{C}$ ),  $t$  is time (s),  $Q$  is the heat source/sink term. The procedure to find out the value of respective terms can be found out in the reference (Burkan Isgor *et al.* 2004). The boundary conditions at concrete surface were found by considering total radiation absorbed, re-radiation by concrete, heat loss by convection and heat conducted into the concrete body. In this study, the thermal conductivity,  $k$ , and specific heat,  $c$ , which depend on the moisture content of the medium and degree of carbonation, was kept constant. The variations in their values were not considered to be significant for normal temperature range.

In a study by (Shafei *et al.* 2012) the equation 5 was solved for a concrete member in a Finite Element framework. The developed finite-element model of the concrete member was subjected to the transient thermal analysis using the ANSYS program. To introduce the initial and boundary conditions, ambient temperature data was gathered for the specific region where the RC member was located. The daily temperature data of the

study area was obtained for the last 15 years from the database. A sinusoidal curve, equation 6, was fitted to the data:

$$T_{env}(t) = 29.1 - 15 \sin\left(\frac{2\pi t}{365}\right) \quad (6)$$

Where, time,  $t$  is in days ( $0 \leq t \leq 365$ ). The boundary conditions were applied to the top and bottom surfaces of the concrete member. It was assumed that the temperature at the top surface follows the trend of equation 6 over time. The bottom surface of the member also has the same trend with the half variation amplitude. Based on the discussed assumptions and equations, the transient thermal analysis is performed on the finite-element model to calculate the temperature distribution in the nodes and elements of the concrete member. The temperature data was stored at each time step and then used as initial conditions for the next time step of the analysis.

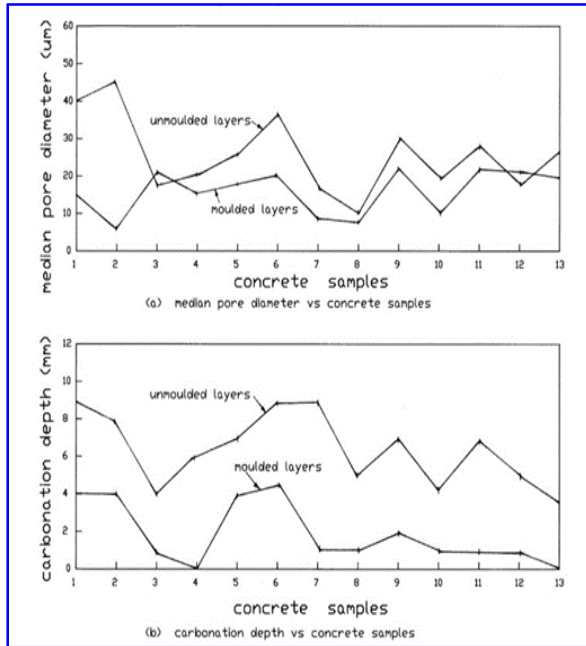
### 3.3 Carbonation and Porosity of Concrete

As carbonation proceeds from the outer surface into internal portions of concrete, the microstructure is also changed continuously from the outer surface into internal portions of concrete (Song *et al.* 2007) and consequently changing the porosity of the concrete. As  $\text{CaCO}_3$  occupies a greater volume than  $\text{Ca(OH)}_2$ , which when it replaces, the porosity is reduced with the advancement of carbonation (Papadakis *et al.* 1991b; Burkan Isgor *et al.* 2004). Higher porosity gives rise to more carbonation, this has been verified experimentally in a study by (Valcuende *et al.* 2010). Natural carbonation of self-compacting concrete was investigated in this study. Eight different Concretes were used which consisted of four self-compacting concrete (SCC) and four normally-vibrated concrete (NVC). The carbonation rate was found to be lower in SCC than NVC. It was attributed to the fact that limestone fines produce less porosity and finer microstructure of concrete. Also, the difference between both types of concrete tended to disappear as their fines content becomes similar.

In a study by Roy (Roy *et al.* 1999) the relationship between carbonation and nature of the pores in the concrete was studied in detail. Larger pores gave rise to higher carbonation depths. Twenty concrete panels were cast and cured for 28 days and kept in two different locations for 2 years. Upon completion of the process, the median pore diameter was determined. Fig. 2 shows that the median pore diameters for the unmolded samples (with large carbonation depth) are greater than those of the moulded samples (smaller carbonation depth). The result presented in the figure 2 which compose the carbonation depth with the median pore size and show a higher carbonation depth for increased pore size is also expected since a higher porosity will produce a higher diffusion rate for carbon dioxide. The importance of uniform and well-compacted concrete is also demonstrated since badly

compacted or honeycombed concrete leads to high porosity and rapid carbonation.

The pore blocking due to rain etc. in concrete also influence the carbonation and hence it should also be accounted in a concrete porosity model. The depth profiling of carbonates formed during natural carbonation of mortars with one face exposed directly to rain and opposite face sheltered has been measured and compared in (Houst et al. 2002). Due to pore blocking by periodic rainwater, the amount of carbonates formed on mortars sheltered from rain was found to be generally higher. As expected and explained in (Papadakis et al. 1991a).



**Fig 2: Median pore diameter and carbonation depth against concrete samples for moulded and unmoulded layers, Source: (Roy et al. 1999)**

A practical method to find initial porosity of concrete can be described as in (Song et al. 2007). The permeability coefficient in carbonated concrete was derived by applying a capillary pore structure formation model in carbonated cement mortar and assuming that aggregates do not affect carbonation process in early-aged concrete as a function of porosity. The permeability obtained from the micro-level modelling for carbonated concrete was verified with the results of accelerated carbonation test and water penetration test in cement mortar. In this study, the permeability coefficient was obtained by the Darcy's Law and the water flow in the unit time given by equation 7

$$\frac{Q_f}{dt} = -\frac{A'dH}{dt} = \frac{K_p HA}{L} \quad (7)$$

$$-\frac{dH}{H} = \frac{K_p A dt}{A'L} \quad (8)$$

Where,  $Q_f$  is total water flow,  $Q_f/dt$  is flow rate,  $A$  is the area of cement mortar,  $A'$  is the area of pipette attached to upside cell,  $L$  is thickness of specimen,  $K_p$  is permeability coefficient,  $H$  is the water head. By integrating equation 8 from initial water head,  $H_0$ , to measured water head,  $H_1$ , as equation 9, the permeability coefficient  $K_p$  was obtained as equation 10.

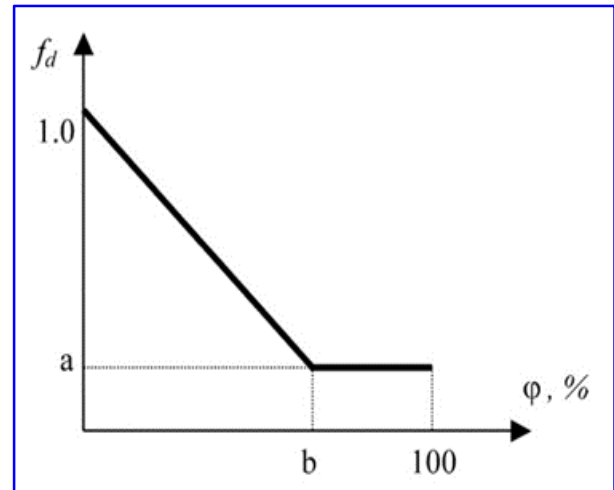
$$-\int_{H_0}^{H_1} \frac{dH}{H} = K_p \int_0^t \frac{A dt}{A'L} \quad (9)$$

$$K_p = \frac{A'L}{At} \ln \frac{H_0}{H_1} \quad (10)$$

The relationship between carbonation and reduction in porosity has also been studied by (Papadakis et al. 1991b), and for a fully hydrated concrete, is given as equation 11.

$$\mu(t) = f_d(t, \varphi) \mu_0 \quad (11)$$

Where,  $\mu(t)$  is the porosity of concrete at time  $t$ ,  $f_d(t, \varphi)$  is a decay function which may be used to determine the changed porosity of concrete due to carbonation,  $\varphi$  is the degree of carbonation,  $\mu_0$  is the initial porosity of concrete. A bi-linear decay function, as in fig was used to simulate the effect of carbonation on the porosity. The parameters  $a$  and  $b$  in can be empirically determined. These are based on observed change in porosity due to carbonation.



**Fig 3: Decay function as a function of degree of carbonation by (Papadakis et al. 1991b)**

Also, initial porosity of concrete  $\mu_0$  can be calculated as equation 12:

$$\mu_0 = \frac{\frac{w}{c} \frac{\rho_c}{\rho_w} (1 - \mu_{air})}{1 + \frac{w}{c} \frac{\rho_c}{\rho_w} + \frac{a}{c} \frac{\rho_c}{\rho_a}} + \mu_{air} \quad (12)$$



Where, for a given concrete,  $a/c$  is the aggregate–cement ratio,  $w/c$  is the water–cement ratio,  $\mu_{air}$  is the air content of the concrete,  $\rho_c$ ,  $\rho_a$  and  $\rho_w$  are the densities of cement, aggregates and water, respectively.

### 3.4 CO<sub>2</sub> Diffusivity

Bjorn Lagerblad (Lagerblad *et al.* 2005) stated that diffusion coefficient depends on microclimatic factors, essentially RH inside the cement paste and therefore, calculation of CO<sub>2</sub> uptake largely depends on laboratory testing and empirical data, including measurements on real concrete structures.

The transport of CO<sub>2</sub> inside concrete is governed by following equation 13 (Burkan Isgor *et al.* 2004) which is analogous to equation 5.

$$\frac{\partial}{\partial x} \left( D_c \frac{\partial C_c}{\partial x} \right) + \frac{\partial}{\partial y} \left( D_c \frac{\partial C_c}{\partial y} \right) + Q_c = \frac{\partial C_c}{\partial t} \quad (13)$$

Where,  $D_c$  ( $m^2 s^{-1}$ ) is the CO<sub>2</sub> diffusion coefficient,  $C_c$  ( $kg m^{-3}$  pore solution) is the CO<sub>2</sub> concentration and  $Q_c$  ( $kg m^{-3} s^{-1}$ ) is a sink term representing the reduction in the CO<sub>2</sub> content of concrete due to carbonation reaction (as discussed in section 2). Papadakis (Papadakis *et al.* 1991b) proposed  $D_c$ , equation 14, as a function of several factors such as temperature, relative humidity, age of concrete, exposure conditions, etc. equation 14

$$D_c = 1.64 \times 10^{-6} \mu_p(t)^{1.8} (1-h)^{2.2} \quad (14)$$

Where,  $h$  is relative humidity, expressed as a fraction, at time  $t$  and  $\mu_p(t)$  is the porosity of the hardened cement paste. Above expression has been used by (Burkan Isgor *et al.* 2004) for their model for prediction of concrete carbonation.

Another model proposed by Shafei (Shafei *et al.* 2012) used the equation, equation 15 for calculation of CO<sub>2</sub> diffusion coefficient. They considered effect of major influential factors such as ambient temperature  $F_{B1}(T)$  relative humidity  $F_{B2}(H)$ , age of the concrete  $F_{B3}(t_e)$ , and change of the concrete pore structure during the carbonation process  $F_{B4}(R)$  and the reference CO<sub>2</sub> diffusion coefficient  $D_{B,ref}$ .

$$D_B = D_{B,ref} F_{B1}(T) F_{B2}(H) F_{B3}(t_e) F_{B4}(R) \quad (15)$$

The values suggested by (Saetta *et al.* 1995) were used for calculation of different terms in equation 15. The reference CO<sub>2</sub> diffusion coefficient for a normal strength concrete in the standard temperature and relative humidity of 65% can be calculated as equation 16:

$$D_{B,ref} = D_{B0} 10^{-0.05 f'_{cm}} \quad (16)$$

Where,  $f'_{cm}$  is the mean value of the 28-day compressive strength of the concrete (MPa) and  $D_{B0}$  is equal to  $10^{-6.1} m^2 s^{-1}$ .

$$F_{B2}(H) = (1-H)^{2.5} \quad (17)$$

Where,  $H$  is the corresponding pore relative humidity.

$$F_{B1}(T) = \exp \left[ \frac{E}{R} \left( \frac{1}{T_{ref}} - \frac{1}{T} \right) \right] \quad (18)$$

Where,  $R$  is the gas constant ( $kJ (mol. K)^{-1}$ ) and  $E$  is the activation energy of diffusion process ( $kJ mol^{-1}$ ). The value of  $E$  for a cement paste made of OPC depends on the water to cement ratio.

$$F_{B3}(t_e) = \chi + (1-\chi) \left( \frac{28}{t_e} \right)^{0.5} \quad (19)$$

Where,  $t_e$  is the age of the concrete (day) and  $\chi$  is the ratio of the diffusivity at the time of diffusivity to the diffusivity after 28 days. And,

$$F_{B4}(R) = 1 - \varepsilon R \quad (20)$$

Where, the value of  $\varepsilon$  ranges between 0 and 1. If the diffusion process results in no reduction in porosity,  $\varepsilon$  would become zero.  $R$  is the degree of carbonation. The degree of carbonation is determined as the ratio of accumulated CO<sub>2</sub> already reacted with Ca(OH)<sub>2</sub> to the maximum possible concentration of reacted CO<sub>2</sub> in the concrete. Further details about these parameters can be read in details in reference (Shafei *et al.* 2012).

### 3.5 Carbonation of Concrete Containing Blended Cements

Blended cements are widespread in use, and different types of cement have very different microstructural and physical properties. Thus, it is essential to understand the nuances of carbonation properties of different hardened cement paste made up of pozzolana cement, ground granulated blast furnace slag cement etc. The effect of supplementary cementing materials (SCM) on concrete resistance against carbonation and chloride ingress has been studied by (Papadakis, 2000). The silica fume and low and high calcium fly ash were used as SCM. Experimental investigations simulating main deterioration mechanisms in reinforced concrete were carried out. It was observed that the carbonation depth decreases as Aggregate replacement by SCM increases and increases as Cement replacement by SCM increases.

#### 3.5.1 Carbonation of Cement Containing Fly Ash

It has been reported that the normal dosages of fly ash addition result in the slight increment of the carbonation of concrete whereas in concrete with normal moist-curing periods, carbonation is not affected by the significant amount. Many studies have reported that higher carbonation has been found in concrete containing fly ash (Byfors *et al.* 1985; Ho *et al.* 1987, 1988; Thomas *et al.* 1990; Papadakis *et al.* 1992; Atiş *et al.* 2003; Sulapha *et al.* 2003). Contrary to these studies, in a case, where a 25-year-old fly ash foundation

concrete was examined, and it has been reported that there has been no carbonation in the fly ash concrete specimens. (Cabrera and Woolley, 1985).

The study by (Thomas *et al.* 1992), concludes that concretes with up to 30% fly ash carbonated to a similar or slightly greater degree compared with OPC concretes of the same strength grade. However, concretes containing 50% fly ash carbonated at significantly greater rates. This study reports carbonation of fly ash concrete, with particular emphasis on the role of curing. It also observes that in some cases increasing the initial curing period from 1 to 7 days had the effect of reducing carbonation by 50%. Concretes with nominal strength grades M25, M35 and M45 and a range of fly ash levels (0-50%) were exposed to various treatments and environment during the first 28 days. After 28 days the concrete cubes were stored either internally or externally (sheltered) and the rate of carbonation was monitored.

Also, Papadakis (Papadakis *et al.* 2000) reported that in the case fly ash is introduced as a fine aggregate replacement, the carbonation rate is reduced. (Khunthongkeaw *et al.* 2006) However, reaction products between pozzolanic silica and  $\text{Ca}(\text{OH})_2$  result in the denser structure of the HCP, so diffusivity is reduced, and carbonation is likely to be slowed down. For this to happen, good curing is essential (Parrott *et al.* 1987; Hobbs *et al.* 1988)

### 3.5.2 Carbonation of Concrete Containing Silica Fume

A study by (Kulakowski *et al.* 2009) signifies the conclusions regarding the depth of carbonation, carbonation induced reinforcement corrosion in concrete samples with silica fume addition upto 20%, and the water/binder ratios ranging from 0.30-0.80. The water-binder ratios specify the behaviour of the additions. In the materials having w/b ratios lower or equal to 0.45-0.50, the porosity of the material is the governing factor in the process of carbonation whereas the consumption of the calcium hydroxide has an insignificant effect on the carbonation. On the contrary for higher w/b ratios, a substantial role is played by the consumption of calcium hydroxide. At the same time, the results concluded from the reinforcement corrosion studies, direct that the outcome of the silica fume addition is to be governed by their concentration. Silica fume will not lessen the corrosion resistance for the concentrations equal to or lower than 10%, and it might actually increase it when used in the concentrations below this level. Silica fume increases the potential for carbonation-induced reinforcement corrosion when used in the concentrations greater than 10%.

### 3.5.3 Carbonation of Concrete Containing Alkali-Activated Blast-Furnace Slag Mortar (AAS)

Different studies on Alkali-Activated Blast-Furnace Slag Mortar (AAS) have shown that AAS as lesser resistance to carbonation (Song *et al.* 2014; Cabrera and Woolley, 1985). Unlike OPC, the

compressive strength of AAS reduces this can be attributed to the fact that, the major hydration product of AAS is calcium silicate hydrate (CSH), with almost no portlandite, unlike the products of OPC. After carbonation, the CSH of AAS turned into amorphous silica gel. And hence, reducing the compressive strength. Also, the increase of the activator dosage leads AAS to react more quickly and produce more CSH, increasing the compaction, compressive strength, and carbonation resistance of the microstructure. (Song *et al.* 2014).

### 3.5.4 Carbonation of Concrete Containing Granulated Blast Furnace Slag Cement (GBFSC)

G. G. Litvan and A. Meyer (Litvan *et al.* 1986) carried out a long-term study to illustrate the comparison of carbonation between granulated blast furnace slag cement (GBFSC) concrete and ordinary Portland cement (OPC) concrete. It was found that the rate of carbonation in GBFSC significantly surpasses, when compared to the rate in OPC concrete. Declination has been shown in the porosity of OPC concrete whereas, in case of GBFSC, it remained same. The basic source of the increase in the permeability of GBFSC concrete with carbonation was the coarsening of the pores and a large decrease was suffered by the tensile strength of the surface region.

Although lots of experimental work has been done, research work performed on the carbonation prediction of the concrete containing SCMs are rare.

Jiang (Jiang *et al.* 2000) developed a mathematical model, equation 21, to predict carbonation depth of high-volume fly ash (HVFA) concrete. In this study, an accelerated carbonation test was carried out on ordinary Portland cement (OPC) concrete and HVFA concrete.

$$X_c = 83.9 (1 - RH)^{1.1} \sqrt{\frac{\left(\frac{W}{B^*} - 0.34\right) C_o}{\alpha k' C}} \sqrt{t} \quad (21)$$

Where,  $W/B^*$  = effective w/c ratio,  $\alpha$  is the degree of hydration,  $C_o$  = concentration of  $\text{CO}_2$ ,  $C$  = cement content,  $k'$  = coefficient related to carbonation reactivity depending on the contents of cement and fly ash,  $n$  is a parameter related to the pore system of concrete and reported to have a value between 2.0 and 2.1. The predicted depth results were compared with the experimental results. Researchers also pointed out that the effective water/binder ratio and the cement content are the important parameters influencing HVFA concrete carbonation.

Wang (Wang *et al.* 2009) suggested a multi-component concept based numerical model which can predict the carbonation of concrete containing silica fume. This numerical model had two parts: hydration and carbonation models. The hydration model takes the mix proportion of concrete as input and considers Portland cement hydration and pozzolanic activity. From the model, the amount of hydration product that is

susceptible to carbonate along with porosity was obtained as a function of curing age. The diffusivity of  $CO_2$  in concrete was also determined and the carbonation depth of concrete was also predicted. The prediction results showed a good agreement with the results of the experiment performed in this study.

### 3.6 Effects of Water/Cement Ratio

The water/cement (w/c) and water/binder ratio are considered as one of the most important design parameters affecting the concrete quality. It has been verified by several studies (Fattuhi *et al.* 1986; Roy *et al.* 1999; Chaussadent *et al.* 2000) that depth of carbonation increases with increase in water/cement ratio. This is attributed to the strong influence of w/c ratio on microstructural properties of concrete. The effect of the water-cement ratio of cement pastes on microstructural characteristics related to carbonation process was studied in (Chaussadent *et al.* 2000). Investigations were performed during the hydration process at an early age and on 28-day and 2-year old hydrated cement pastes. The mix was made with a Type I Normal Portland Cement. The w/c ratios used in this study were 0.25, 0.35, 0.45, and 0.60. It was observed that the microstructural characteristics are very different between pastes having w/c values above 0.35-0.40 and below 0.35-0.40. When w/c is more than this value, the porosity strongly increases, and the calcium hydroxide appears in large crystals. It was also observed that, for an equivalent degree of hydration, the calcium hydroxide amount increases and the CaO/SiO<sub>2</sub> ratio of the C – S – H decreases as the w/c increases.

An experimental study by (Roy *et al.* 1999) to understand the effect of water-cement ratio on carbonation has been conducted, using w/c ratios of 0.55, 0.60, 0.65, 0.70 and 0.75. The cubes were cast, and their compressive strength was measured after 28 days of curing, another prism cast of the same sample was cured for 7 days and transferred to the carbonation chamber and their carbonation depths were measured weekly, for 6 weeks. The data of carbonation depth are summarized in Table.6 It has been observed that samples having higher w/c ratios, i.e. the ratios, which are of a lower grade of concrete shows larger carbonation depths. The rate constants, K (mm/year<sup>0.5</sup>) were calculated on the basis of carbonation depths, assuming carbonation is a diffusion-controlled phenomenon and the carbonation depth d (in mm), is correlated to the exposure time, t (years) by the equation 22:

$$d = K \sqrt{t}^{0.5} \quad (22)$$

The calculated K-values are shown in Table 6: it has been observed that K increases with increasing w/c ratio or decreasing cube strength. Similar relationship between concrete strength (grade, quality) and carbonation rate has been quite well established in several previous studies.

I was concluded that there exists a relationship between the rate (depth) of carbonation and the strength of the concrete sample. The carbonation depth was found to be proportional to (strength)<sup>-1</sup> and the carbonation rate constants, K (mm year)<sup>0.5</sup> measured in the accelerated laboratory tests are significantly higher than those found in normal atmospheric conditions.

Similar results were also obtained in the study by (Fattuhi *et al.* 1986). Accelerated carbonation technique used to investigate the effects of concrete mix w/c ratio on the rate of concrete carbonation. Prisms 50 x 50 x 285 mm made from concretes with w/c ratios of 0.7, 0.6 or 0.4. The prisms were initially water cured for 1, 3, 7, 21 or 28 days; then they were placed in a chamber filled with CO<sub>2</sub> gas. The carbonation depth was examined using phenolphthalein indicator. The results showed that the depth of carbonation increased with increase in the concrete w/c ratio.

### 3.7 Effect of Ambient Humidity and Moisture Content

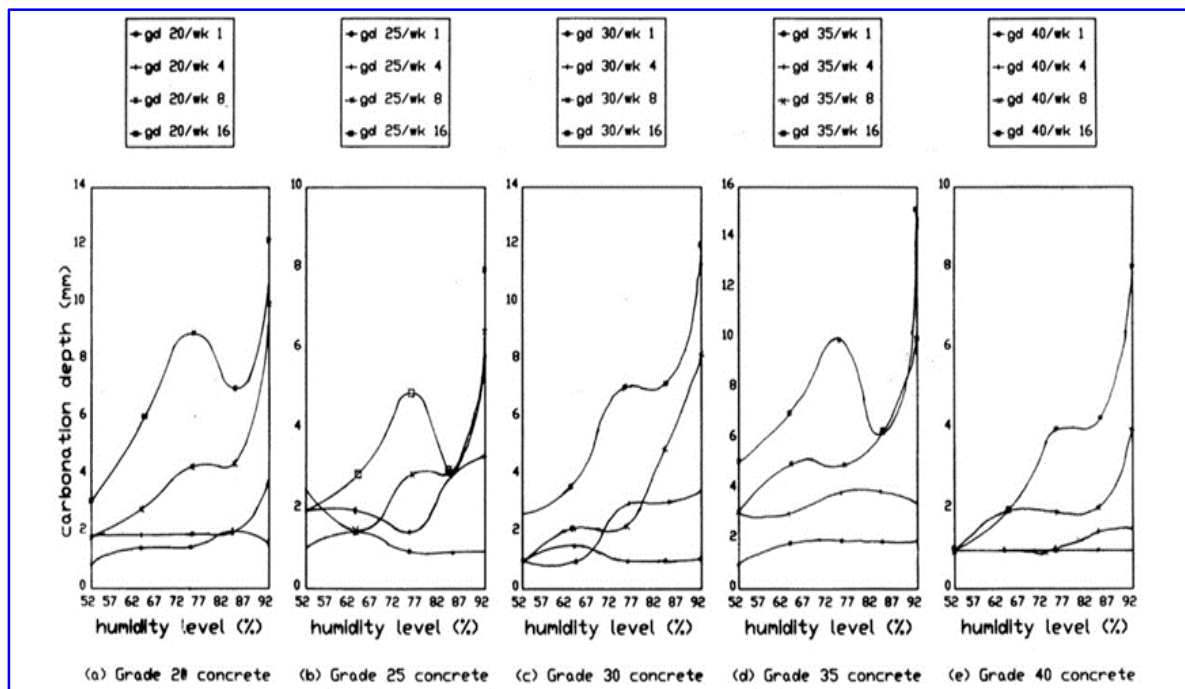
Deterioration of the concrete may be caused by various chemical processes, and water may act as an essential component in these reactions (Burkan Isgor *et al.* 2004). For lower relative humidity (RH less than 50%), diffusion of CO<sub>2</sub> into concrete is high, but there may not be enough water in the pores to support carbonation. For a high RH, diffusion of CO<sub>2</sub> is very low, which leads to a reduction in carbonation and its carbonation rate (Papadakis *et al.* 1991b; Papadakis *et al.* 1992; Castel *et al.* 1999) That is why the majority of the studies on concrete carbonation use RH between 50% and 70%.

Roy (Roy *et al.* 1999) conducted an experimental study to find the effect of humidity levels on the depth of carbonation, the author used five different grades of concrete (grade 20, 25, 30, 35 and 40 with w/c ratio of 0.8, 0.7, 0.65, 0.6 and 0.55 respectively). After curing for 28 days, the samples were transferred in special chambers having CO<sub>2</sub> at 6% by volume with humidity levels 52, 64, 75, 84 and 92%, to accelerate the carbonation process. Carbonation tests were carried out at five different exposure periods viz. 0, 1, 4, 8 and 16 weeks.

From Fig. 4, it is visible that significant carbonation is visible only after 8 or 16 weeks. By looking at the carbonation depth v/s humidity level relationship after 16 weeks exposure, the same general trends can be seen from all five grades of concrete. As the humidity level increases from 52 to 75%, there is a significant increase in carbonation depth with increasing humidity level. There is then a decrease in carbonation depth as the relative humidity increases from 75 to 84% before the carbonation depth increases once again as the relative humidity increases to 92%.

**Table 6: Cube Strength, Carbonation Depths and Carbonation Rate Constants K, for the Grades of Concrete**

Water/cement ratio	Cube strength (Mpa)	Carbonation depth (mm) after different exposure periods						Carbonation rate constant K (mm/year <sup>-0.5</sup> )
		7	14	21	28	35	42	
0.55	27.0	0	1	2.2	3.0	3.7	5.3	10.89
0.60	26.5	0	1	2.8	3.3	4.5	5.5	11.89
0.65	23.5	0	1	3.3	3.8	5.1	7.1	13.7
0.70	20.5	0	1	3.9	4.3	6.1	7.5	15.5
0.75	18.5	0	1	4.1	4.7	6.3	8.5	16.9

**Fig. 4: Carbonation depth v/s humidity level for different grades of concrete (Roy et al. 1999)****Table 7. Details of mix used in (Russell et al. 2001) (NOTE: Fine aggregate/course aggregate ratio held constant at 0.55)**

Mix no.	w/c ratio	Cement content kg m <sup>-3</sup>	Water content kg m <sup>-3</sup>	Fine aggregate content kg m <sup>-3</sup>	Course aggregate content kg m <sup>-3</sup>	Achieved a/c ratio
1	0.50	375	192	685	1243	5.14
2	0.57	375	215	664	1207	4.99
3	0.63	375	234	647	1174	4.86
4	0.70	375	256	625	1140	4.71
5	0.50	315	164	732	1329	6.54
6	0.57	315	184	711	1296	6.37
7	0.63	315	201	696	1296	6.23
8	0.70	315	220	679	1233	6.07

So, based on the experimental study, it can be concluded that the effects of RH were complex with maxima being measured at both 75% and 92% RH. The former maxima are similar to real structures and the enhanced carbonation at latter maxima RH may be

possible due to the difference in the mechanism (s) of carbonation in the accelerated depths (Roy et al. 1999).

Author, (Russell et al. 2001) conducted a study to investigate the effect of relative humidity on the rate of carbonation. Based on data from an



accelerated carbonation test, descriptive models were developed to quantify the effects of the relative humidity on the rate of carbonation. Eight different concrete mixes were used. Four different w/c ratios and two cement contents (CC) were used. Three batches of these eight mixes were cast for testing at three different RH conditions: 55, 65 and 75% RH. All other properties like curing, conditioning and exposure to the  $CO_2$  environment was held constant for all mixes. Details of the eight mixes used in the test are given in Table 7. Class 42.5N Portland cement (OPC) conforming to BS12: 1991 (BS12: 1991) was used. The carbonation chamber was set to maintain a 5%  $CO_2$  ( $\pm 0.5\%$ ) concentration at  $20^\circ C$  ( $\pm 1^\circ C$ ) during the testing. Carbonation was measured using 1% phenolphthalein solution. Internal humidity was monitored at 0, 10, 20, 30- and 40-mm depths from the exposed surface.

The rates of carbonation of all mixes are given in Table 8 using the equation 22. The overall trend observed is that rate of carbonation decreases with increasing RH. For the lower w/c ratio mixes, the greatest change in the rate of carbonation occurred between 55% and 65% target RH. Maximum carbonation occurred at 55–65% RH. From 65 to 75% RH there was a decrease in the rate of carbonation.

**Table 8: Carbonation rates obtained for different values of RH in (Russell et al. 2001)**

w/c ratio	Cement content kg m <sup>-3</sup>	Target relative humidity		
		55%	65%	75%
		Carbonation rate mm/(week <sup>0.5</sup> )		
0.50	375	3.32	1.43	1.97
0.57	375	5.56	2.90	2.39
0.63	375	5.49	6.08	3.06
0.70	375	9.84	7.94	2.74
0.50	315	4.40	1.71	1.32
0.57	315	4.48	4.20	2.42
0.63	315	8.72	6.34	2.82
0.70	315	10.96	7.62	4.18

#### 4. CONCLUSION

A review has been presented with the aim of examining the effects of different parameters influencing carbonation of concrete. Further, uncertainties, similarities and dissimilarities across different experimental studies has been discussed. Also, insights have been given on different analytical and empirical models developed so far. The following findings are reported:

I. Carbonation is a multivariable and nonlinear phenomenon. The factors influencing carbonation of concrete are interdependent, and the entire process is highly complex. Thus, modelling for prediction of concrete carbonation should be based on nonlinear scheme. Nonlinear FEM based

models performed satisfactorily for carbonation depth prediction. Several neural network-based models have also been developed. Because of nonlinear and interdependent nature of the problem ANN, GA and other artificial intelligence-based models can be deployed effectively.

II. Equation 2, which defines a linear relationship between carbonation depth and the square root of time with a proportionality constant, has been used by several researchers for reporting the effects of influencing parameters on carbonation of concrete. This equation considers diffusion properties to be constant which does not reflect real scenario. Also, as observed across different experimental data the proportionality-constant changes appreciably. Although, highly convenient in comparing results in controlled laboratory conditions but this equation should not be used to study or predict long-term natural carbonation of concrete.

III. Carbonation resistance of concrete increases with increase in curing period. Different curing regimes do not affect carbonate on until it causes a change in the characteristic strength of concrete. Curing period of three days is sufficient for developing good carbonation resistance in concrete.

IV. The high-temperature environment was found to be favourable for the formation of the passivated surface of rebars in the SPSs. But the dissolution velocity of the passivated surface is also bigger in the high-temperature SPSs. Thus, the rebars have the greater capacity of passivity at a lower temperature.

V. In general, carbonation of concrete causes the increase in the thermal conductivity and all mechanical properties of concrete like strength, modulus of elasticity, and shrinkage of the concrete.

VI. Higher porosity gives rise to higher carbonation depths. To ensure lower porosity or denser structure, concrete should be uniform, well compacted and has high cement content. Carbonation causes a reduction in porosity of the concrete. Therefore, carbonation slows down with time or its propagation. Pore blocking due to rain may also influence carbonation propagation. Hence, in a porosity model, a function which incorporates initial concrete properties as well as other variables like degree of carbonation, relative humidity, temperature etc. must be used.

VII. In general, the carbonation depth decreases as aggregate replacement by SCM increases and increases as cement replacement by SCM increases. Concretes with up to 30% fly ash carbonated to a similar or slightly greater degree

compared with OPC concretes of the same strength grade. However, concretes containing 50% fly ash were found to carbonate at significantly greater rates.

- VIII. The rate of carbonation was found to be increasing with increase in w/c ratio. Water-Cement ratio is the most important parameter influencing carbonation of concrete. Concrete having lower w/c ratios were found to be more carbonation resistant even with smaller curing periods.
- IX. As the humidity level increases from around 50% to 75%, there is a significant increase in carbonation depth with increasing humidity level. There is then a decrease in carbonation depth as the relative humidity increases from 75 to 84%.

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